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Published in:
European Physical Journal E

DOI:
10.1140/epje/i2001-10099-1

Publication date:
2002

Citation for published version (APA):
Weaire, D., Cox, S., & Fátima Vaz, M. (2002). The transition from two-dimensional to three-dimensional foam structures. *European Physical Journal E*, *7*(4), 311-315. https://doi.org/10.1140/epje/i2001-10099-1

Eur. Phys. J. E **7**, 311–315 (2002)
DOI 10.1140/epje/i2001-10099-1

THE EUROPEAN PHYSICAL JOURNAL E
EDP Sciences

The transition from two-dimensional to three-dimensional foam structures

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Received 15 October 2001 and Received in final form 14 January 2002

Abstract. Small cells in an experimental sample of two-dimensional foam, such as that which is contained between two glass plates, may undergo a transition to a three-dimensional form, becoming detached from one boundary. We present the first detailed observations of this phenomenon, together with computer simulations. The transition is attributed to an instability of the Rayleigh-Plateau type. A theoretical analysis is given which shows that an individual cell is susceptible to this instability only if it has less than six sides.

PACS. 47.20.Dr Surface-tension-driven instability – 82.70.Rr Aerosols and foams

1 Introduction

In recent years, both 2D and 3D foam structures have come to be well understood, with the help of various programs for simulation [1–4].

In the 2D case, comparison is made with the structure obtained by trapping bubbles between two glass cells (Hele-Shaw cell), or between one plate and a liquid surface [5,6]. In either case, the system is not strictly two-dimensional: the assumption of two-dimensionality, in both experiments and numerical simulations, must rely on criteria such as the transverse thickness being less than, or the radius of curvature greater than, the bubble size. The second procedure makes it straightforward to vary the transverse thickness of the system, as in the experiments reported here and recent investigations of multiple layers of monodisperse foam [7].

The present paper is addressed to the transition to a 3D structure which must occur as the thickness is increased. We shall see that this is usually triggered by an instability, closely related to the Rayleigh-Plateau instability of a fluid cylinder.

Experimental results are presented for the onset of the instability in the case of a cluster of seven bubbles, consisting of a central cell surrounded by six others, which we shall call "petals". The results are found to be consistent with theoretical estimates based upon a Rayleigh-Plateau–type analysis and with computer simulations using the Surface Evolver.

Transitions of this kind have occasionally been noted in the grain growth of thin films, which is a closely related although not identical problem, being a matter of kinetics rather than static equilibrium. In that case, the vanishing of a small cell must bring it into the regime of the Rayleigh-Plateau instability identified here, so that a 2D/3D transition is likely before it disappears entirely. In what follows we shall consider only the equilibrium structures, although our numerical calculations show intermediate structures which assist in interpreting the evolution of the cell-shapes; these should not be viewed as dynamic calculations.

Our own concern with this transition arose in the context of a theory of strictly 2D instabilities [8]. Attempts to confirm these by experiment are sometimes frustrated by the prior occurrence of the type of instability considered here.

We shall first review the standard theory of the Rayleigh-Plateau instability, and then introduce the corresponding instability for a cluster of cells. Experiments on such a cluster are described; they show that the thickness at which the instability occurs and the final arrangement of the bubbles both depend upon the relative volumes of the bubbles.

2 The 2D/3D transition for a single bubble

Consider first the case of a single bubble of volume V trapped between two parallel plates whose separation is d. For small d there is a stable solution in the form of a cylinder. When d is increased to the critical value $d_{\mathrm{crit}} = \sqrt[3]{V\pi}$ this solution becomes unstable [9], and proceeds, via a distortion in the shape of a wine bottle, to detach from

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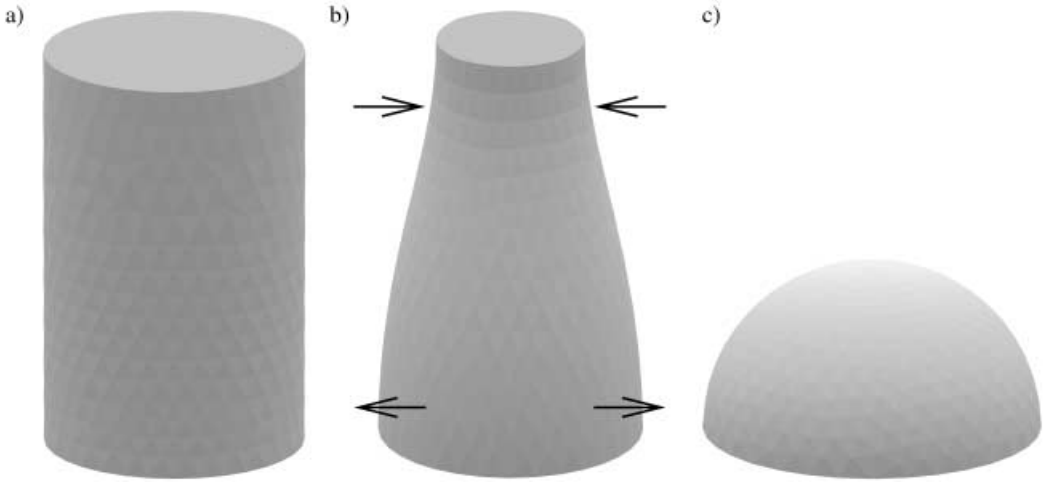

Fig. 1. The Rayleigh-Plateau instability for a single bubble, simulated using the Surface Evolver. a) A cylinder of volume $V = 1$ is stable when the separation between the plates is $d = 1.45$. b) When d is increased to $d = 1.47$ the instability is seen, as one end of the bubble starts to shrink. (Note that this is not a true dynamical calculation; the picture shows an intermediate structure in the process of area minimisation after the instability is encountered.) c) The bubble detaches from, in this case, the top plate to form a hemisphere attached to the lower plate.

one of the containing boundaries, as shown in Figure 1. This is a familiar instability of the Rayleigh-Plateau type, with the longest wavelength compatible with the boundary conditions at the two ends. The second longest wavelength corresponds to a distortion having the shape of an hourglass.

The standard theory of this surface-tension–driven instability is given by [9]. We prefer to couch the argument in terms of a bubble's cross-sectional area A and perimeter L, since this leads more naturally to our argument in the case of a cluster of cells.

Consider a volume-preserving perturbation applied to a cylinder which has initial perimeter $L_0 = 2\pi R_0$ and cross-sectional area $A_0 = \pi R_0^2$. The surface energy is proportional to the surface area, which varies with height z:

$$S = \int_0^d L(A(z))\sqrt{1+\left(\frac{\mathrm{d}R}{\mathrm{d}z}\right)^2}\,\mathrm{d}z = \int_0^d L(A(z))\sqrt{1+\frac{1}{4\pi A}\left(\frac{\mathrm{d}A}{\mathrm{d}z}\right)^2}\,\mathrm{d}z \quad (1)$$

and since $\mathrm{d}A/\mathrm{d}z$ is small, we can split this into two terms:

$$S \approx \int_0^d L(A(z))\mathrm{d}z + \int_0^d \frac{L(A(z))}{8\pi A}\left(\frac{\mathrm{d}A}{\mathrm{d}z}\right)^2\mathrm{d}z = S_1 + S_2. \quad (2)$$

A Taylor expansion of S_1 gives

$$S_1 \approx \int_0^d L_0\mathrm{d}z + \int_0^d \frac{\mathrm{d}L}{\mathrm{d}A}(A-A_0)\mathrm{d}z + \frac{1}{2}\int_0^d \frac{\mathrm{d}^2L}{\mathrm{d}A^2}(A-A_0)^2\mathrm{d}z. \quad (3)$$

The first term is constant, as for an unperturbed cylinder, and volume preservation implies that the second term is always zero. However, the second derivative of the perimeter L with respect to area A is negative, so that the contribution of S_1 is to lower the surface energy by introducing this variation of cross-sectional area with height. It is this that drives the instability.

Counteracting this effect is an increase of area due to the finite slope of the function $A(z)$. This corresponds to the expression S_2, which is positive for any finite perturbation. The balance of these two terms results in a minimum wavelength for instability with respect to sinusoidal perturbation. The critical separation d_{crit} is half of that wavelength.

This is the most elementary example of the 2D/3D transition. One example of its counterpart for a cluster of cells is shown in Figure 2. This was found using the Surface Evolver software package [2]. In the following sections this more complicated transition is explored in terms of experiment, simulation and theory.

3 Experimental results

We studied the stability of a 2D cluster consisting of a central cell of volume V_{c} surrounded by six "petals" of equal volume V_{p}, as shown in Figure 2a. The petals were produced by blowing air through a capillary immersed in a detergent solution, as in previous work on 2D foams [5, 6], and trapped between the level of the solution and a glass plate [10]. A further bubble of volume V_{c} is added by blowing air through a syringe and the plate is tilted and re-leveled [10] to reassemble the cluster with the central bubble in the middle.

The transition to a 3D cluster is initiated by changing the height d of the cluster, *i.e.* the separation between the

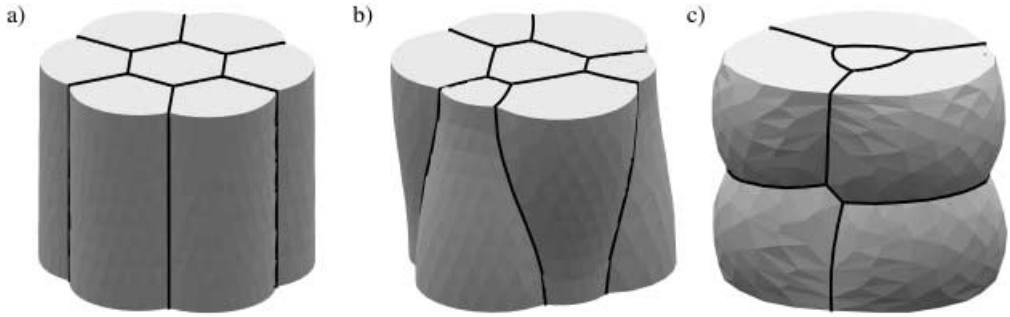

Fig. 2. The Rayleigh-Plateau instability for a simulated cluster of seven bubbles of equal volume $V = 1$. a) A central cell is surrounded by six others, and is stable with $d = 1.8$. b) When the separation is increased to $d = 1.83$, the central cell remains stable, but each of the "petals" exhibits a "wine-bottle" instability, which takes the cluster towards the state shown in c). This latter configuration is again an intermediate state before one of the arrangements shown in Figure 4 is attained.

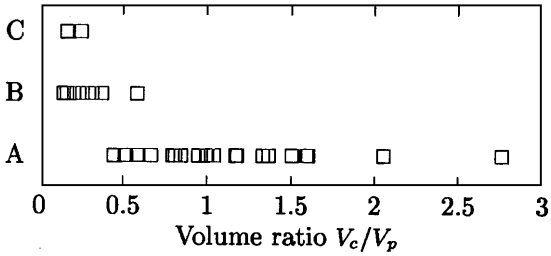

Fig. 3. When the instability occurs, one of three possible configurations of the bubbles is found. We plot the occurrence of each of the arrangements A, B and C as a function of the ratio of bubble volumes V_c/V_p. They are illustrated in Figure 4. For $V_c > V_p$ configuration A is always obtained, whereas for small V_c/V_p we see either B or C.

liquid surface and the top plate, by removing liquid from the vessel. At a certain separation $d = d_{crit}$ the instability takes place and two layers of cells are formed.

Four sets of experiments were performed in which the volume of the petals V_p was kept constant while V_c was varied. In each of the cases $V_p = 15, 18, 47$ and 56 mm^3 we measured d_{crit} with a precision of 0.5 mm and found it to be the same for each value of V_p. Therefore we conclude that the formation of a second layer depends only upon the volume of petals and not on the volume of the inner cell. Moreover, it is the petals that drive the instability.

The arrangement of the seven bubbles in the consequent 3D cluster can vary. We obtained three types of configurations A, B and C (pictured in Fig. 4) depending on the value of V_c/V_p, graphed in Figure 3. The experimental data do not allow us to define precise intervals for the occurrence of each configuration. However, we can state that for $V_c > V_p$ configuration A is found whereas for $V_c < \frac{1}{2}V_p$ the bubbles arrange to form B or C. Configuration C is very rare: it was obtained only twice in 40 experiments and appears for very small values of V_c/V_p, for which configuration B is sometimes produced. This suggests that the energy of 3D clusters with configurations B or C should be close.

4 Simulations using the Surface Evolver

We have used the Evolver to reproduce the 3D clusters that were observed in the experiments, after the instability had occurred. Figure 4 shows configurations A, B and C. In each case the petal volumes are $V_p = 1$, and we have varied the central volume in accordance with the relative volumes observed in each configuration in the experiments. For the purposes of illustration the separation is $d = 1.4$, though this is not necessarily the critical value of d.

The surface energies of these clusters, each with petal volume $V_p = 1$ and central volume $V_c = 0.5$, are as follows (for unit surface tension): $E_A = 20.02, E_B = 19.97$ and $E_C = 20.22$. Thus we would expect configuration B with these bubble volumes. For a larger central bubble volume, $V_c = 1.5$, we find $E_A = 21.65, E_B = 22.31$ and $E_C = 22.15$, indicating that configuration A should be found for $V_c > V_p$.

We have made further use of the Surface Evolver to study the stability of clusters with different numbers of petals and with different bubble volumes. For the cluster with six petals (Fig. 2) we start with a very thin ($d = 0.2$) configuration, and slowly increase d to seek the instability, moving in increments of $\Delta d \approx 0.1$ and converging at each step. At $d = d_{crit} = 1.85$ we see the petals deform into the wine-bottle shape, as shown in Figure 2b. As a bubble starts to shrink from one plate, both of its neighbours shrink from the opposite plate. To pinpoint d_{crit} it is necessary to decrease d slightly and then increase it more slowly. This value of d_{crit} is independent of the volume of the central bubble, in agreement with the experimental observation that it is the volume of the petals that determines the critical separation.

Following the instability to its conclusion using the Surface Evolver is difficult, due to the change in topology when a bubble separates completely from a bounding surface. Our calculations suggest that the cluster evolves to the configuration shown in Figure 2c. This is unstable however, and the final configuration of the seven-bubble cluster is one of either A or B. It would seem that configuration C will only occur on those occasions when just two pairs of petals become unstable.

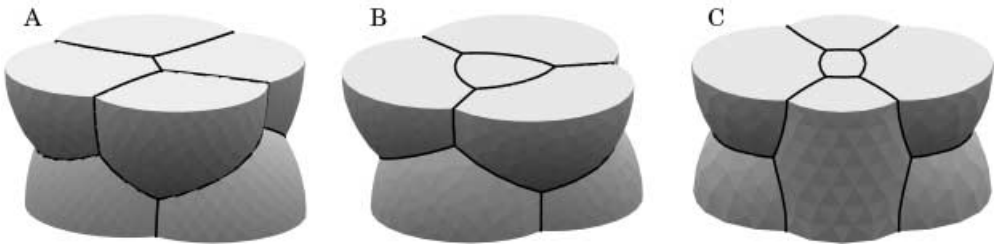

Fig. 4. Possible configurations for seven bubbles simulated using the Surface Evolver, with plate separation $d = 1.4$ and six bubbles of unit volume. Configurations A, with $V_c = 1.5$, B ($V_c = 0.5$) and C ($V_c = 0.5$) are reproduced from the experimental observations. A and B both have four bubbles touching the upper plate and three on the lower one. In configuration C there are three bubbles that touch both plates. These are the three possible final states of a cluster of seven bubbles that has undergone the 2D to 3D transition.

We have applied the same method to a cluster with only four petals surrounding the central cell. The response, as d is increased, depends upon the relative volumes. If the central bubble has a lower volume than the petals, $V_c < V_p$, we find that it is the central bubble that drives the instability. It detaches from one of the plates and moves to the other plate. For a large central bubble, $V_c \geq V_p$, it is again the petals that deform due to the wine-bottle instability.

5 Theory of the Rayleigh-Plateau instability for a foam cell

The Rayleigh-Plateau or wine-bottle instability may affect small cells in any foam which is nominally 2D but has a finite transverse dimension, such as the typical experimental sample of 2D soap froth. The number of sides of the cell in question is crucial: only cells with less than six sides are, according to the following argument, susceptible to this instability, however small they become. It is, of course, precisely such cells that shrink to zero size in the coarsening process [4] so we expect each of these vanishing cells to finally undergo the transformation provoked by such an instability. (In practice, however, effects associated with the finite size of the junctions at vertices may also play a role.)

We consider only a fully symmetric foam cell in a cluster of N cells, and analyse the effect of its shape and connections based upon the standard treatment of a cylindrical (single bubble) cell given earlier.

The (central) foam cell has n vertices, area A_0, perimeter L_0, and for convenience we define [11]

$$k_n = \frac{L_0}{\sqrt{A_0}}. \tag{4}$$

For such symmetric cells, k_n varies little with n, being within a few percent of 3.71 for all n [11]. In the standard treatment of Rayleigh-Plateau, the instability is driven by a term proportional to the *negative* second derivative of the perimeter with respect to the area. It is opposed by a positive term that is proportional to the perimeter, and the two are equal at the critical point for instability.

In modifying this, it must be borne in mind that it is the total perimeter of the foam or cluster that is involved in the first term, that is, the change of perimeter with area of the neighbouring cells is to be included. The effect of this is that the second derivative of perimeter with respect to area can be negative or positive, mainly depending on the number n' of sides. Of course, $n = n'$ except for the special case $n = 0, n' = 1$.

It is clear from scaling that

$$E(A_0) = E(0) + \sqrt{A_0} f(n). \tag{5}$$

We could proceed to evaluate $f(n)$ from geometrical considerations, but recall instead that we can express the first derivative of the energy with respect to A_0 as the pressure difference between the cell and its neighbours. This is closely connected with the well-known theory of the Von Neumann Law [4]. The theorem of Gauss which equates the integral of turning angle to 2π for a closed cell allows this to be written as

$$\frac{\mathrm{d}E}{\mathrm{d}A_0} = \frac{\gamma}{L_0}\frac{\pi}{3}(6-n) = \frac{\gamma}{k_n\sqrt{A_0}}\frac{\pi}{3}(6-n)\,, \tag{6}$$

where γ is the surface tension in the films. Hence the corresponding approximation for the second derivative is

$$\frac{\mathrm{d}^2E}{\mathrm{d}{A_0}^2} = -\frac{\pi\gamma}{6k_nA_0^{3/2}}\,(6-n). \tag{7}$$

For a finite number of sides, the factor of $6-n$ is introduced (together with other less significant modifications). For $n > 6$ the second derivative of E with respect to A_0 reverses sign. Recall that it is the negative sign of this quantity that drives the instability, so this is suppressed for $n > 6$. Although we have not yet analysed the case of a non-symmetric cell, it would seem that this result should not be strongly affected.

Recall that d_{crit} is determined by the balance between the driving force S_1 and the positive term S_2. The latter remains positive for the case of a foam cell, and of roughly the same magnitude. It follows that a foam cell of given volume V having n sides, with $n < 6$, should undergo the Rayleigh-Plateau instability at a value of transverse thickness d given *approximately* by

$$d_{\mathrm{crit}}(n) = d_{\mathrm{crit}}(1)\sqrt{\frac{6}{6-n}} = \sqrt[3]{V\pi}\sqrt{\frac{6}{6-n}}. \tag{8}$$

Table 1. The theoretical prediction of critical thickness $d_{\rm crit}$, from equation (8), of a specified bubble in a cluster of N bubbles is compared with simulation (upper table) and also with experiment in the cases included in the lower table. To allow comparison with the (averaged) experimental values, $d^*_{\rm crit}$ is calculated by dividing the asterisked value in the upper table by $\sqrt[3]{V_{\rm p}}$. There is semiquantitative agreement throughout.

N	$V_{\rm c}$	$V_{\rm p}$	Bubble	Number of vertices	$d_{\rm crit}$ simulation	$d_{\rm crit}$ theoretical
1	1	–	central	0	1.47	1.46
5	1,2	1	petal	4	1.94	2.54
5	0.5	1	central	4	1.68	2.01
7	0.5,1,1.5	1	petal	4	1.83^*	2.54
7	0.5,1,1.5	1	central	6	stable	stable

N	$V_{\rm c}$ (cm^3)	$V_{\rm p}$ (cm^3)	Bubble	Number of vertices	$d_{\rm crit}$(cm) experiment	$d^*_{\rm crit}$ simulation	$d_{\rm crit}$ theoretical
7	0.009–0.03	0.0147	petal	4	0.45	0.45	0.62
7	0.004–0.05	0.018	petal	4	0.55	0.48	0.66
7	0.008–0.12	0.047	petal	4	0.65	0.66	0.92
7	0.014–0.077	0.056	petal	4	0.60	0.70	0.97

This estimate is for a single cell undergoing such an instability by itself. It should be suitably modified in cases in which neighbouring cells undergo simultaneous instabilities, as is indeed the case in some of our preliminary experiments.

6 Comparison of theory, experiment and simulation

Let us apply this theory to the experiments and simulations described earlier. Our results are summarised in table 1. For the cluster with six petals, the theory correctly predicts that it is always the petals that drive the instability, since they have four sides, while the central bubble has six and is thus stable. The critical value of d obtained from the simulations is $d_{\rm crit}(4) = 1.83$. The value predicted by equation (8) is $\sqrt[3]{\pi}\sqrt{3} = 2.54$, which is an overestimate, as might be expected due to the asymmetry of the petals. A similar degree of overestimation is found with the experimental data in the seven-bubble case. However, the agreement between the simulated $d_{\rm crit}$ and the (averaged) experimental values is excellent.

For the cluster of five bubbles, each of the bubbles now has four sides. The simulations suggest that the relative volumes are important in deciding whether it is the central bubble or the petals which first becomes unstable. This is reflected in the values for $d_{\rm crit}$ given by the theoretical prediction equation (8), which shows that for any four-petal cluster in which the central bubble has a volume less than that of the petals, $V_{\rm c} < V_{\rm p}$, it is the central bubble which will become unstable first.

We can extend this result to any cluster of N bubbles with $N-1$ petals. For $N \geq 7$ it is always the petals that are unstable, while the central bubble is stable, since it has six or more sides. For smaller clusters, $3 \leq N \leq 6$, the central bubble should become unstable first if

$$\frac{V_{\rm c}}{V_{\rm p}} < \left(\frac{7-N}{2}\right)^{3/2}. \qquad (9)$$

This formula can easily be adapted to the case of a more extensive polydisperse foam.

The Surface Evolver software package can also calculate the Hessian H, that is, the matrix of second partial derivatives of the energy. This has been used, for example, to study the stability of two liquid drops which are squeezed together [12] or of two-dimensional bubble clusters [8]. In further computational work we will use the Hessian to further elucidate the mechanism of these transitions.

The authors wish to thank F. Graner and M.A. Fortes. This research was supported by the Prodex programme of ESA. SJC acknowledges funding from a Marie Curie fellowship.

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